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Composition and Distribution of Polynuclear Aromatic Hydrocarbons Contamination in Surficial Coastal Sediments from Odidi Area of Delta State, Nigeria

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Abstract: *The distribution of PAHs in the environment has become widespread due to the impacts of human activities especially those relating to oil exploration and exploitation. This study was carried out to determine the composition and distribution of PAHs in sediments from Odidi River, Delta State, Nigeria. Samples were collected from two (2) upstream and downstream sampling points each at depths of 0 - 15cm from March 2015 to August 2016. Gas chromatography-mass spectrometry was used to determine the poly nuclei aromatic hydrocarbons (PAHs) concentrations in the samples. All data analyses were done using Microsoft Excel 2016. The results obtained showed that the mean concentrations of PAHs ranged from 4.37±1.29µg/g to 23.13±1.42µg/g (upstream), and from 2.22±1.11µg/g to 25.00±2.63µg/g (downstream). There was no significant difference between the values obtained upstream and downstream. Across the 18 months of the study, only Florene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benzo(a)anthracene and Crysene were detected in every month. The composition of PAHs shows a higher concentration of Heavy PAHs (HPAHs) with the mix of Light PAHs (LPAHs) indicating that they were released by both petrogenic and pyrogenic sources in the study area. The PAHs content of the sediments exceeds the threshold effect concentration (TEC) TEC and the threshold effect levels TEL limits PAHs in sediments indicating potential environmental and health threats.*

Keywords*: PAHs, Sediments, Odidi river, Niger Delta region, oil pollution*

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1.0 Introduction

Polyaromatic hydrocarbons (PAHs), also known as polycyclic aromatic hydrocarbons or poly-nuclei aromatic hydrocarbons, are hydrocarbon compounds which are composed of aromatic ring molecules that are fused (Ihunwo *et al.*, 2021). Each ring in the structure shares at least one side with another and contains delocalised electrons. The PAHs contain atoms of carbon and hydrogen only (Sahoo *et al.*, 2020). Common examples of PAHs are naphthalene, coronene, pentacene, chrysene, pyrene, phenanthrene and anthracene. They are generally non-polar and lipophilic, and these properties make them environmentally persistent due to their poor solubility in water. Large PAH molecules are solids and in their pure forms exhibit pale green, pale yellow and white colours, or may be colourless (Ali *et al.*, 2022). PAHs are formed from natural and man-made sources. Volcanic eruptions and forest fires are major natural sources, while petroleum and coal contain PAHs in abundance. Human activities including incomplete combustion of fossil fuels and wood, high-temperature cooking, combustion of waste and cigarette

smoking are all sources of PAHs in the environment (Patel *et al.*, 2020).

The entry of PAHs into environmental media occurs through processes including runoff, wastewater treatment, petroleum spillage and atmospheric deposition. When released into the environment, PAHs contaminate the air and water bodies while becoming bound to sediments and biota even after the removal of the source (Zhang *et al.*, 2021). They contribute to the depletion of the ozone layer Aquatic organisms exposed to these compounds exhibit toxic, mutagenic, carcinogenic and teratogenic effects. The photosynthetic processes in exposed plants are disrupted leading to inhibited growth and development (Ali *et al.*, 2022).

PAHs are important human health factors due to their roles in diseases and genetic damage. This is further compounded by their long residence time in environmental media. Exposure occurs through air, food and dermal contact (Mallah *et al.*, 2022). Acute toxicity of PAHs in humans is low, however, chronic exposure to PAHs, particularly in the occupational setting, results to increase in the occurrence of cancers in the bladder, skin and lungs (Barbosa Jr. *et al.*, 2023). The carcinogenic PAHs are benzo[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, chrysene, dibenzo(a,h)anthracene and indeno(1,2,3-cd)pyrene (Singh *et al.*, 2020). Non-carcinogenic impacts of PAHs are also seen in the dermal, renal, gastrointestinal and pulmonary systems. These include cardiovascular diseases, higher occurrence of obstructive lung problems, worsening asthma and decreased lung function (Ma *et al.*, 2022). Due to the occurrence of PAHs as mixes, and the fact that effects occur after long-term, low-level exposure, the best method to protect human health is to gain knowledge of factors that increase exposure risk including inhaling smoke, consuming charred meats and handling petroleum products (Mosallaei *et al.*, 2023). Therefore, this study was carried out to assess the composition and distribution of PAHs in coastal sediments in Odidi River, Delta State, Nigeria.

2.0 Experimental methods

2.1 Study area

Odidi River is located in Warri South-West Local Government Area of Delta State, Nigeria. It is situated within the Niger Delta region of the country and lies at latitude 5°26'18" N and longitude 5°27'37" E (Fig. 1). People who inhabit these areas are majorly involved in fishing. Oil exploration and production activities are common in the area as it is home to facilities such as flow stations, gas plants and oil fields owned by oil-producing companies in Nigeria. Areas near the water body have been consistently polluted by petroleum products since the first discovery of oil in the region (Ogeleka *et al.*, 2016).

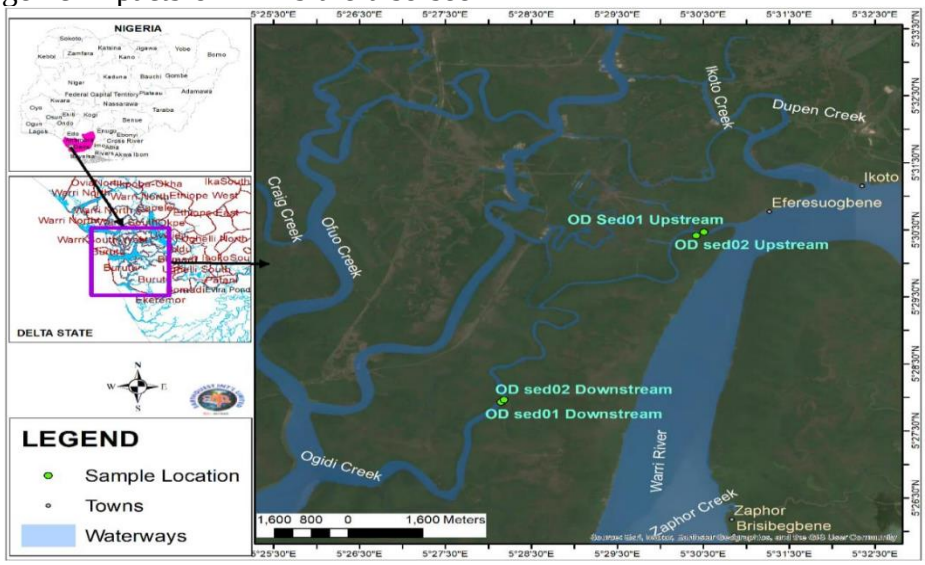

Fig. 1: Map of the study area and sites for samples collection

2.2 Sample collection and preparation

Surface sediment samples were collected in the coastal region of Odidi, Delta State. Four sampling points were selected (that is two upstream and two downstream) and an Ekman sampler was used to collect sediments at 0 - 15cm depths. For each point, 5 subsamples were taken and mixed well producing a homogenous sample. This sampling took place monthly from May 2015 to August 2016. Sediment samples were air-dried for 72 hours and then stored in the laboratory before analysis.

2.3 Analytical procedures

The concentration of PAHs in the sample was determined following the method described by Jasiewicz *et al.* (2007). Extraction of 5g of the sample was carried out using 10 mL of acetonitrile in an ultrasonic bath for 60 minutes. The mixture was set down for 10 minutes and filtered. Purification of the extract was done using column chromatography incorporating CHROMAFIX 400-SA cartridge solid phase extraction to extract the fractions of 16 PAHs in the sample. Three mL methanol was used to condition the CHROMAFIX 400-SA cartridges. One millilitre of the sample was passed through the cartridge and methanol was used to elute the PAHs. High-purity nitrogen was used to concentrate the extracts to 1mL through a gentle stream. Gas chromatography-mass spectrometry was used to determine the concentrations of 16 PAHs (naphthalene, acenaphthalene, acenaphthene, florene, phenanthrene, Anthracene, fluoranthene, pyrene, benzo(a)anthracene, crysene, benzo (b) fluoranthrene, benzo(a)pyrene, benzo(k) fluoranthene, Indeno(1,2,3) perylene, dibenzo(a,h)anthracene and Benzo(g,h,i)perylene) on an Agilent GC 6890/MS/MS 5973 apparatus having an ion trap. The resolution of PAHs was attained by a Factor Four DB-5MS capillary column (length 30 m, internal diameter 0.25 mm, film thickness 0.25 μm). Helium with a flow rate of 1mL/min was the carrier gas, while the transfer line and injector temperature was 325 °C. The programming of the GC oven was: 40 °C for 2 min, followed by a 30 °C min^{-1} ramp to 250 °C and then with a ramp 10 °C min^{-1} to a final temperature of 320 °C (2.5 min). The ion source temperature for the mass spectrometer was 150°C and its electron energy was 70eV. The detector was operated in SCAN mode with a scanning range of 70 - 300m/z. PAH identification in the sample was done by creating a match of the mass spectra of each PAH and its retention time with that of the standard. The standard used was Restek "610 PAH Calibration Mix B" (16 PAHs in methylene chloride: methanol (1:1, v:v, 100–2000 μg/mL)) (Jasiewicz *et al.*, 2007).

2.4 Statistical analyses

Microsoft Excel Sheets 2016 were used in the analysis and presentation of data. Mean, standard deviation and ANOVA were calculated for the data.

3.0 Results and Discussion

3.1 Spatial distribution

PAHs in the samples were all above 2 μg/g as shown in Table 1.

Aside from some of the samples in April 2015. The mean values for upstream samples ranged from 4.37±1.29μg/g to 23.13±1.42μg/g, while those of downstream samples ranged from 2.22±1.11μg/g to 25.00±2.63μg/g (Fig. 2).

Across the entire study period, the overall total PAH content of the downstream sampling points was slightly higher than that of the upstream sampling points. Both the maximum and minimum recorded PAH values were from the upstream portion of the river.

The highest total upstream PAH content was recorded in August 2015 while the lowest in April 2015. For the downstream section, the highest concentration was also recorded in August 2015 but the lowest was in July 2015.

Out of the 16 PAHs detected, only florence, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene and crysene were recorded every month.

Table 1: Total concentrations of PAHs for each month

Month	µg/g					
	Upstream			Downstream		
	1	2	Mean±SD	1	2	Mean±SD
March 2015	21.66	12.25	16.96±4.70	20.1	14.8	17.46±2.66
April 2015	11.63	1.48	6.55±5.07	3.3	1.1	2.22±1.11
May 2015	6.09	6.92	6.50±0.42	5.2	6.1	5.62±0.45
June 2015	4.92	9.68	7.30±2.38	21.4	16.1	18.75±2.68
July 2015	3.08	5.66	4.37±1.29	2.1	12.0	7.06±4.97
August 2015	21.71	24.56	23.13±1.42	22.4	27.6	25.00±2.63
September 2015	12.78	16.15	14.47±1.68	10.1	7.3	8.71±1.38
October 2015	10.52	6.20	8.36±2.16	10.7	10.7	10.71±0.01
November 2015	8.47	9.05	8.76±0.29	12.3	11.5	11.93±0.39
December 2015	5.62	8.48	7.05±1.43	3.6	9.1	6.37±2.75
January 2016	11.11	8.24	9.67±1.44	11.7	5.9	8.80±2.90
February 2016	6.11	8.53	7.32±1.21	7.3	12.2	9.73±2.43
March 2016	12.75	14.28	13.51±0.77	22.3	11.1	16.70±5.64
April 2016	4.51	14.32	9.41±4.91	13.6	10.8	12.19±1.42
May 2016	6.34	4.09	5.22±1.12	3.9	3.1	3.46±0.40
June 2016	15.43	12.56	13.99±1.44	13.5	3.0	8.266±5.236
July 2016	16.94	19.16	18.05±1.11	17.5	21.6	19.50±2.05
August 2016	9.43	11.19	10.31±0.88	15.0	12.7	13.82±1.14

This contrasted with Ekere *et al.* (2017) where only 6 PAHs were detected in similar sediments from the confluence of rivers Niger and Benue. The PAH concentrations differed widely from those of other studies from the Niger Delta region. The results recorded exceeded those reported by Arowojolu *et al.* (2021) where total PAHs in similar sediments from the River Niger ranged from 0.06 - 0.38µg/g. However, the values were similar to Iwegbue *et al.* (2021a) who reported PAHs concentrations of Rivers Forcados, Ase and Niger to be 1.62 - 19.80 µg/g, 2.93 - 16.10µg/g and 2.40 - 19.00 µg/g.

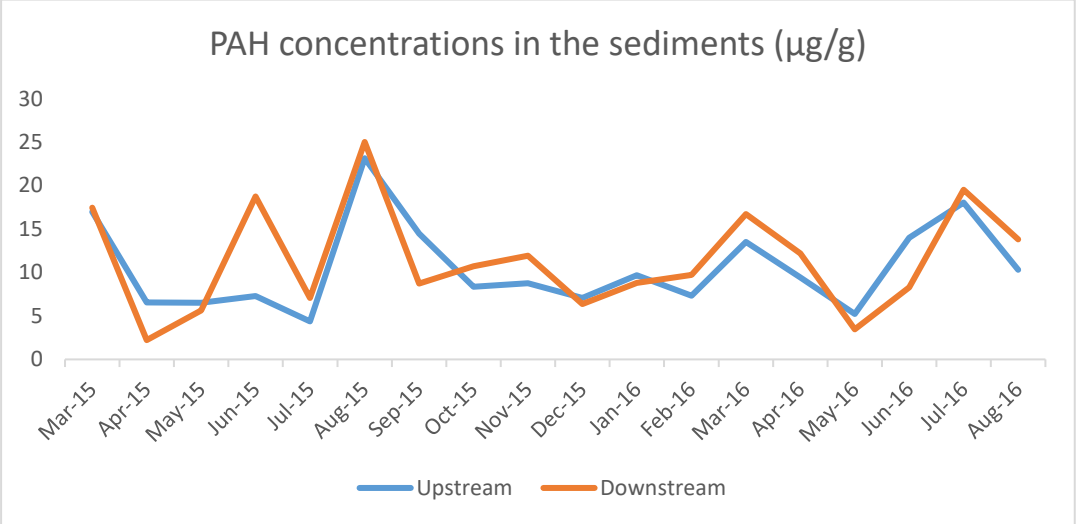

Fig. 2: Comparative mean concentrations of PAHs in the sediments

Similarly, Benson *et al.* (2020) reported that PAH concentrations in sediments of Lagos lagoon ranged from 1.43 - 5.90µg/g. Parra *et al.* (2020) also, reported similar concentrations of PAHs in sediments with values ranging from 0.49 - 5.62µg/g. However, the results were less than that of Inam *et al.* (2018) where PAH concentrations in Ikpa River was 1.05µg/g. Also well documented is the PAH concentrations by Iwegbue *et al.* (2021b) from Escravos River which ranged from 0.75 - 213µg/g. Furthermore, Li *et al.* (2023) reported PAH concentrations ranging from 0.098 - 0.239µg/g in Bohai and Laizhou bays, China, which were far higher than the results of this study. Even higher levels of PAHs were recorded by Baran *et al.* (2017) in sediments taken from nine reservoirs in Poland - Rybnik, Rzeszów, Brzóza Królewska, Brzóza Stadnicka, Besko, Chechło, Ożanna, Głuchów and Narożniki with concentrations ranging from 150 - 33,900µg/kg in nine reservoirs located in Poland.

The results of ANOVA for concentrations of PAH upstream and downstream in the study area showed that there was no significant difference ($p>0.05$) in concentrations (Table 2).

Table 2: ANOVA for PAH concentrations

	SS	df	MS	F	P-value	F crit
Between Groups	6.547628	1	6.547628	0.209632	0.649968	4.130018
Within Groups	1061.951	34	31.23387			
Total	1068.499	35				

NB: SS = sum of squares; df = degree of freedom; MS = mean square

PAHs in sediments generally pose threats to benthic organisms (McGrath et al., 2019). The threshold effect concentration (TEC) for total PAHs in sediments was reported as 2900µg/kg, that is, 2.9 µg/g, while the threshold effect level (TEL) was reported as 1684µg/kg, that is, 1.684µg/g (McGrath et al., 2019). The values of total PAHs recorded in this study mostly exceeded these limits and should be a source of concern for environmental health sake.

3.2 PAHs composition

PAHs are grouped into two: light PAHs (LPAHs) and heavy PAHs (HPAHs).

The LPAHs are Naphthalene, Acenaphthalene, Acenaphthene, Fluorene, Phenanthrene and Anthracene, while the HPAHs are Fluoranthene, Pyrene, Benzo(a)anthracene, Crysene, Benzo (b)fluoranthrene, Benzo(a)pyrene, Benzo(k)fluoranthrene, Indeno(1,2,3)perylene, Dibenzo(a,h)anthracene and Benzo(g,h, i)perylene (Mou *et al.*, 2023). Fig. 3 and 4 show the compositions of PAHs in the samples based on the above classification. Fig. 3 shows the upstream PAH compositions. It revealed that the HPAH content was higher than that of LPAH for 12 of the 18 months that the study period covered. Fig. 4 showed that for the downstream section and revealed that HPAH content exceeded that of LPAH in 13 of the 18 months. In the upstream samples, the LPAH ranged from 0.00 - 16.83µg/g while the HPAH ranged from 1.19 - 15.06µg/g. On the other hand, in the downstream, the LPAH ranged from 0.46 - 10.09µg/g and the HPAH ranged from 0.00 - 20.34µg/g.

The dominance of heavy PAHs in the samples was observed. Benson *et al.* (2020) reported that generally, sediments in Lagos Lagoon and Ikpoba River, Benin City, both in Nigeria were dominated by HPAHs with concentrations ranging from 1.31 - 4.43µg/g, while LPAHs ranged from 0.12 - 2.57µg/g in Lagos Lagoon. However, Areguamen *et al.* (2023) reported that sediment samples contained more PAHs having 3 (LPAH) and 6 rings (HPAH) in Ikpoba River, Nigeria, across the period of their study. However, the dominance of PAH content by LPAH was

reported by Li *et al.* (2023) where they accounted for 56% of the total PAH content in Laizhou and Bohai Bays, China.

The dominance of HPAHs in the samples indicates nearby sources of pollution which in this case, could be attributed to the oil exploration activities which has been on in the vicinity (Mou *et al.*, 2023). The presence of HPAHs in the sediments was a pointer to potentially higher environmental and health risks compared to LPAHs which have lower persistence in the environment (Areguamen *et al.*, 2023).

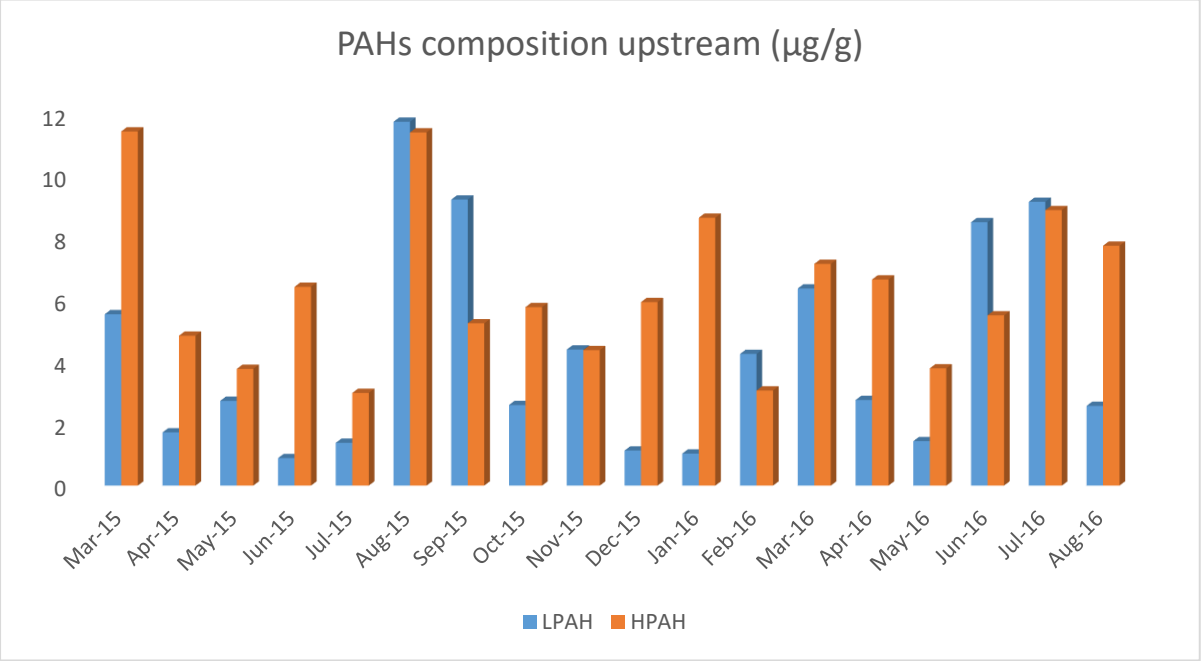

Fig. 3: PAHs composition in upstream samples

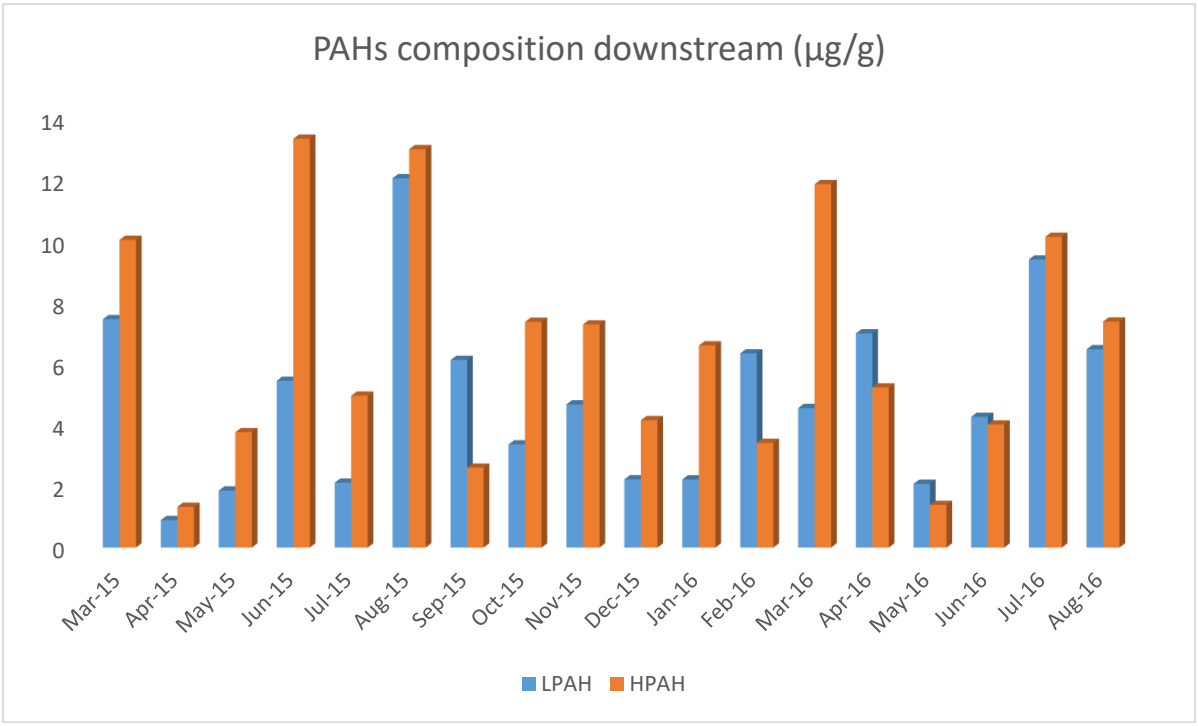

Fig. 4: PAHs composition in downstream samples

Furthermore, the composition of PAHs in the samples indicated that they came from a mix of petrogenic and pyrogenic sources. Petrogenic sources of PAHs in the study area include oil spills and other use of petroleum products not related to combustion, while the pyrogenic sources include gas flaring and other combustion processes (Patel *et al.*, 2020). This is further buttressed by the finding that the ratios of pyrene/fluoranthene and phenanthrene/anthracene vary widely from less than 1 in some samples to greater than 10 in others.

4.0 Conclusion

This study evaluated the distribution and composition of polynuclear aromatic hydrocarbons (PAHs) in sediments from Odidi area of Delta State, Nigeria. The results show that the PAH content exceeded the TEC and TEL limits recommended for PAHs in river sediments indicating potential environmental threats to benthic organisms and health threats to human consumers of the aquatic biota from the river. Also, it observed that the PAHs composition is dominated by heavy PAHs with light PAHs being lower across all sampling locations. The hydrocarbon indicators also showed that the PAHs in the area are from a mix of petrogenic and pyrogenic sources. The study further revealed that remedial actions are required to protect environmental and human health within the river and by extension similar PAHs polluted aquatic environment.

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Compliance with Ethical Standards Declaration

Ethical Approval

Not Applicable

Competing interests

The authors declare that they have no known competing financial interests.

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Availability of data and materials

Data would be made available on request.

Authors' contributions

Both authors contributed equally to the work.